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"In-situ diffraction and imaging studies of hetero-epitaxial growth of semiconductors"

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ABSTRACT

Our work emphasizes in-situ characterization of the initial stages of heteroepitaxial growth of semiconductors and ultrathin film silicides using advanced instrumentation and techniques, including high resolution reflection high energy electron diffraction (RHEED), a UHV scanning electron microscope with micro-probe RHEED and a UHV scanning transmission electron microscope (UHV-STEM). Systems of interest include vicinal Si(100), germanium on silicon, and ultrathin film silicides.

Specific instrument and technique developments include: Demonstration that Auger lineshapes can be used to separate coexisting silicide phases in a partially reacted ultrathin film; Demonstration that quasi-kinematic RHEED intensity calculations can be used to identify epitaxial structures; Imaging of single atomic height steps with STEM; Visualization of submonolayers of germanium and various metals using biassed secondary electron imaging; Auger imaging at the highest spatial resolution obtained anywhere.

Specific scientific accomplishments include: Measurement of a kinetic phase formation diagram for contact reactions in ultrathin film silicides showing previously overlooked thickness dependence; Identification of Ni2Si-theta as a precursor structure responsible for the much studied B- to A-type transition in NiSi2 overlayers; Demonstration that single layer steps on Si(100) are favored over double layer steps due to strain rather than entropic effects, as previously believed; Identification of novel strained island structures for Ge on Si(100); Determination of surface diffusion and binding energies related to nucleation and growth processes for Ge on Si(100).

OBJECTIVES

The objective is to characterize fundamental aspects of the initial stages of heteroepitaxial growth, with emphasis on microscopic aspects. Specific areas of study include:

1. The thermodynamics and kinetics of atomic steps on Si(100), both clean and with adsorbates, such as arsenic or germanium.
2. Nucleation and growth of Ge/Si(100).
3. Low temperature silicide formation in ultrathin films.

STATUS

A. SCIENTIFIC RESULTS:

1. Atomic steps on Si(100):

Control of atomic steps on Si(100) is fundamental to successful heteroepitaxial growth on silicon, particularly for the suppression of anti-phase boundaries in compound semiconductor overlayers, and for the preparation of low dimensional devices, such as quantum wires and dots. The topic of steps on semiconductor surfaces has attracted a great deal of attention recently from the viewpoint of fundamental physics as well. There is a fascinating interplay of long range strain vs. short range rebonding effects, roughening and other phase transitions involving step edge and kink interactions, and growth kinetics effects. In many ways, Si(100) is a prototype system in which to study these phenomena. To this end, we have developed powerful methods for systematic investigation of step interactions using microprobe RHEED and a lenticular substrate. With this arrangement, we have available, on a single sample, a continuous range of polar and azimuthal miscut angles, with corresponding terrace lengths and inter-kink spacings, respectively.

We have shown that single-height steps, which are deleterious to overlayer growth, are thermodynamically stable for miscut angles of <3deg (terrace lengths of <20Å). The transition from single-to biatomic steps is broad and independent of temperature, which implies that the mechanical energy of step meander is much larger than previously believed (pub 10, 11). This data provides firm numbers for the hotly debated "phase transition" on this surface, and is stimulating much discussion. We plan to extend this work to include the effects of "surfactant overlayers", such as arsenic or antimony, and to compare with the behavior for Ge(100). In addition, we have begun calculations of diffraction lineshapes from which to infer details of step and kink configurations. (ref 12) These calculations include the effects of correlated sublattices (2x1 and 1x2), and dimers, which cannot be treated with existing analytical methods.

2. Nucleation, Growth and Coarsening of Ge/Si(100):

We have made in-situ observations of the nucleation and growth of Ge on vicinal Si(100) at various temperatures and thicknesses, using direct imaging in the biased secondary electron mode in the UHV-STEM (pub.6). The secondary electron images have been used to obtain size distributions, spanning some 4 orders of magnitude in cluster density, of a quality not previously obtained. This work is currently being analyzed in detail and published(pub 6,7). During annealing at 375 and 525C, we find diffusion lengths of many microns, and low sticking coefficients at step and island edges. Anomalous coarsening behavior suggests interesting thickness dependent instabilities in the intermediate layer, and shows the dramatic effects of coherently strained islands. Parameters of growth including nucleation density, surface diffusion lengths and the effects of steps have been described in appropriate atomistic models (pub. 8).

In parallel with the above work, we have been developing the highest spatial resolution Auger electron spectroscopy and imaging available anywhere, and have used Ge/Si(100) as a test sample. We have obtained Auger image resolution of better than 5nm (pub 7,9).

3. Silicide Formation:

We have developed special techniques of RHEED intensity analysis and Auger lineshape analysis for in-situ characterization of ordered and disordered structures, respectively that are particularly suited to study the growth of ultrathin film silicides (pubs 3, 17). This involved development of an auxiliary sample preparation chamber to allow in-situ fracture of bulk silicide "reference" compounds (pub. 4), and implementation of a computer controlled vidicon system (pub. 2). With these special techniques, we have identified numerous precursor, low temperature phases in the growth of nickel and cobalt silicides (pubs 1,3,4,14). These metastable phases may have useful device applications, and they can appreciably affect the growth of high temperature epitaxial silicides. From the kinetic phase formation diagrams we have compiled, one can extract information about the fundamental reaction processes such as whether they are diffusion or nucleation limited. Such information facilitates predictive control over thin film growth, possibly allowing the production or stabilization of new metastable compounds. We have shown this for the pseudomorphic Ni2Si-theta structure on Si(111) (pub. 2). We have also used these methods to study fundamental aspects of silicide formation at room temperature. We found for Ni/Si(111) that growth occurs in the form of layers followed by islands, and progresses through compounds of different stoichiometry. This reaction is "spontaneous", being presumeably driven by the heat of condensation, and appears to be diffusion limited (pubs 5,13). These results have direct bearing on the growth of materials at low temperature by codeposition.

B. EQUIPMENT DEVELOPMENT:

The development of advanced instrumentation and techniques has comprised a substantial portion of this project, as described below.

1. RHEED and MBE system:
We have built a precision RHEED system and associated thin film growth chamber. The main innovation of the RHEED system is the use of double deflection magnetic scanning coils both before and after the sample, which allows full control of diffraction conditions, with a fixed gun and detector system. The RHEED system is functional using a vidicon + computer detector. It is our aim to install an energy filtered detector to suppress inelastic scattering, thus improving the contrast of the signal. This will be particularly useful for measuring short range order in polycrystalline overlayers. This method will be complementary to SEXAFS, since multiple neighbor spacing can be determined, along with information on particle size and orientation and surface roughness. A dual e-gun deposition system with TSP and cryoshroud is constructed, but not yet installed. It is designed for silicide codeposition, and semiconductor alloy (Si-Ge) growth. We also developed miniature e-bombardment sources (silicon, nickel, cobalt) for use pending development of the MBE system.

2. UHV-SEM:
We are working to enhance the capabilities and performance of the SEM system, primarily to achieve single atomic step imaging capabilities. This involves improving the mechanical stability, magnetic shielding and field cancellation, and adding a diffraction aperture system. During this period we have designed a dynamic magnetic field compensation system, which addresses our worst resolution limitation at present. An aperture system has been constructed and installed, but not yet tested.

3. UHV-STEM (code named MIDAS):
This instrument combines UHV technology with a state of the art STEM. AFOSR did not support development of this instrument. However, a number of publications describing it were forwarded to AFOSR on March 21, 1990. AFOSR-supported scientific results from this instrument are reported in pubs. 6-12.

MIDAS has all the standard Vacuum Generators STEM features: field emission gun, bright and dark-field detectors, diffraction pattern recording and electron energy loss (EELS) detectors. In addition there is provision for in-situ surface analysis using secondary and Auger electrons. It has attached UHV specimen preparation chambers that allow sample heating to >1200C, and deposition from an array of

Knudsen cell and e-bombardment sources. The acceptance tests gave exceptional results: MgO lattice fringes (0.148nm) and EELS resolution of <0.7eV. A particularly innovative feature is the incorporation of a "parallelizer" detector for low energy Auger and secondary electrons, which promises very high count rate due to a nearly 2π acceptance angle. The high collection efficiency has been demonstrated in biassed secondary electron images of Ge/Si(100) and Ag/Si(100), and in the first Auger electron spectra and scanning Auger images which have around 5nm resolution (pub 9,10). For our applications, the ability to obtain highly significant size distributions using secondary images taken over a wide magnification range (pub 6,7), to see steps (pub 12) and to do composition analysis with Auger spectroscopy is essential.

PERSONNEL SUPPORTED ON AFOSR #87-0367

1. **P. A. Bennett,** principal investigator.
2. **A. P. Johnson**, graduate research assistant; MS thesis "A Surface Phase Diagram for Thin Nickel Films on Silicon(111)" (Aug. 1987).
3. **J. R. Butler**, graduate research assistant; PhD thesis: "Auger and RHEED studies of silicide formation", (Dec. 1990).
4. **X. Tong**, graduate research assistant; PhD thesis: "Mono- to biatomic phase transition on vicinal Si(100) studied with microprobe RHEED", (est. Mar. 1991).
5. **M. R. Wood**, undergraduate hourly; technical help (mechanical).
6. **R. Tatro**, undergraduate hourly; technical help (computers).
7. **S. Ghosh**, graduate hourly; technical help (mechanical).

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PUBLICATIONS SUPPORTED ON AFOSR #87-0367

1. "A Phase Formation Diagram for Precursors to Epitaxial Growth of NiSi2 on Si(111)," P.A. Bennett, A.P. Johnson and B.N. Halawith, Phys. Rev. B37 (1988) p. 4268.
2. "Thin Film Crystallography Using RHEED 'rod intensity profiles': Ni/Si(111)," P.A. Bennett, X. Tong and J.R. Butler, Jour. Vac. Sci. Tech. B6 (1988) p. 1336.
3. "Ultrathin film growth of silicides studied using microprobe RHEED and Auger" P. A. Bennett, J. R. Butler and X. Tong, Jour. Vac. Sci. Tech. A7 (1989) p. 2174.
4. "A kinetic phase diagram for ultrathin film Ni/Si(111)": Auger lineshape results", Mat. Res. Soc. proc. vol 148 (1989) pp 61-69.
5. "Stranski-Krastonov growth of Ni in Si(111) at room temperature", J. R. Butler and P. A. Bennett, Mat. Res. Soc. proc. vol 159 (1990) pp. 159-166.
6. "Studies of Ge/Si(100) and observation of atomic steps on Si(100) using biassed secondary electron imaging in a UHV-STEM", M. Krishnamurthy, J. S. Drucker

and J. A. Venables, Proc. 12th Intl. Cong. for Electron Microscopy (Seattle) 1, (1990).

7. "Microstructural evolution during the hetero-epitaxy of Ge on vicinal Si(100) J. Appl. Phys, M. Krishnamurthy, J. S. Drucker and J. A. Venables, in prep.
8. "Nucleation and growth mechanisms in hetero-epitaxial films", J. A. Venables, J. S. Drucker, M. Krishnmurthy, G. Raynerd and T. Doust, Mat. Res. Soc. proc. (Spring 1990).
9. "Auger electron spectroscopy with probe size limited resolution", G. G. Hembree, J. S. Drucker, F. C. H. Luo and M. Krishnamurthy, Appl. Physics Lett., in prep.
10. "Secondary and Auger electron spectroscopy and energy selected imaging in a UHV-STEM", G. G. Hembree, F. C. H. Luo and J. A. Venables, Proc. 12th Intl. Cong. for Elec. Microscopy (Seattle 1990) 2, 382-3.
11. "Heteroepitaxy of Ge on vicinal Si(100)", M. Krishnamurthy, J. S. Drucker and J. A. Venables, Mat. Res. Soc. proc. (Spring 1990).
12. "Visualiztion of atomic steps by biassed secondary electrron imaging", J. S. Drucker, M. Krishnamurthy and J. A. Venables, Ultramicroscopy (1990) in prep.
13. "Low temperature silicide formation: Ni/Si", J. R. Butler, J. S. Drucker and P. A. Bennett, Jour. Vac. Sci. Technol. (in prep).
14. "A Phase formation diagram for Co/Si(111): RHEED results", P. A. Bennett, B. Devries and J. R. Butler, proc. Mat. Res. Soc. Fall, 1990 (in prep).
15. "Terrace width induced domain transition on vicinal Si(100) studied with microprobe RHEED", X. Tong and P. A. Bennett, Phys. Rev. Lett. (submitted).
16. "Diffraction lineshapes on vicinal Si(100)", X. Tong and P. A. Bennett, proc. Mat. Res. Soc. Fall, 1990 (in prep).
17. "Numerical Auger lineshape analysis of phase formation in Ni/Si(111)" J. R. Butler, X. Tong and P. A. Bennett, Surf. Sci. (in prep).

PRESENTATIONS SUPPORTED ON AFOSR #87-0367

1. "RHEED studies of thin film formation with a UHV-SEM" **P. A. Bennett** and A. P. Johnson, Wickenburg workshop, Wickenburg, AZ Jan 11, 1988.
2. "Precursor Structures in the Formation of Epitaxial NiSi2/Si(111)" **P. A. Bennett**, J. R. Butler and X. Tong , PCSI-15, Asilomar, CA Feb. 1, 1988.
3. "Thin film growth studies using microprobe RHEED and Auger" **P. A. Bennett**, seminar presented at IBM, Yorktown heights, June 1988.
4. "Thin film growth studies using microprobe RHEED and Auger" **P. A. Bennett**, J. R. Butler and X. Tong, 35th Amer. Vac. Soc. mtg. , Atlanta, GA, Oct. 3, 1988 (**invited**).
5. "Structure identification in thin films using RHEED" **P. A. Bennett**, J. R. Butler and X. Tong, Wickenburg workshop, Wickenburg, AZ Jan 4, 1989.
6. "Compound formation in the Ni/Si(111) ultrathin film system studied with numerical Auger lineshape analysis" **J. R. Butler**, X. Tong and P. A. Bennett, March APS mtg. 1989.

7. "A phase formation diagram for ultrathin film growth of nickel silicides" **P. A. Bennett**, J. R. Butler and X. Tong, Spring Mat. Res. Soc. mtg. April 24, 1989 **(invited)**.
8. "Numerical Auger lineshape analysis of phase formation in Ni/Si(111)" J. R. Butler, X. Tong and **P. A. Bennett**, 36th AVS Symp. Boston, MA. Fall, 1989.
5. "Stranski-Krastonov growth of Ni in Si(111) at room temperature", J. R. Butler and **P. A. Bennett**, Mat. Res. Soc. Fall 1989.
6. "Heteroepitaxy of Ge on vicinal Si(100)", **M. Krishnamurthy**, J. S. Drucker and J. A. Venables, Mat. Res. Soc. Spring 1990.
7. "Nucleation and growth mechanisms in hetero-epitaxial films", **J. A. Venables**, J. S. Drucker, M. Krishnmurthy, G. Raynerd and T. Doust, Mat. Res. Soc. Spring 1990 **(invited)**.
8. "Low temperature silicide formation: Ni/Si", J. R. Butler, J. S. Drucker and **P. A. Bennett**, 37th Amer. Vac. Soc. mtg, Oct. 1990 (submitted).
9. "A Phase formation diagram for Co/Si(111): RHEED results", **P. A. Bennett**, B. Devries and J. R. Butler, Mat. Res. Soc. Fall, 1990 (submitted).
10. "Mono- to biatomic step transition on vicinal Si(100)", **X. Tong** and P. A. Bennett, 37th Amer. Vac. Soc. mtg. Oct. 1990 (submitted).
11. "Diffraction lineshapes on vicinal Si(100)", X. Tong and **P. A. Bennett**, Mat. Res. Soc. Fall, 1990 (submitted).